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SPIN WAVE THEORY OF TWO-SUBLATTICE UNIAXIAL HEISENBERG FERRIMAGNETS WITH EXTERNAL MAGNETIC FIELD I. ENERGY SPECTRA, STABLE MAGNETIC PHASES AND CRITICAL FIELDS

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Using the free-spin-waves approximation (FSWA) the influence of a homogeneous external magnetic field on a two-sublattice Néel-type uniaxial Heisenberg ferrimagnet with positive isotropic and anisotropic exchange interaction between the sublattices is considered. Two special cases are examined, namely, when the field is parallel (longitudinal) and perpendicular (transversal) to the anisotropy axis.

In Part I, the field-dependent approximate ground state of the system is obtained by minimizing the expectation value of the spin Hamiltonian in the class of sublattice saturation states, and the system is shown to have two stable magnetic phases in the longitudinal as well as transversal field cases. The double-branch spin wave energy spectra in each phase are determined by diagonalizing the Hamiltonian in the FSWA by means of Bogoliubov's transformation. In the transversal-field case, the critical field for the transition from the canted-spin to the paramagnetic phase, obtained in an earlier paper from the stability conditions for the ground state, is shown to coincide with that following from examining the reality and positiveness of the energy spectra. In Part II, the dependence of the total and sublattice magnetizations and susceptibilities on the field and temperature is studied in the long-wavelength low-temperature approximation.

1. *Introduction*

Ferrimagnets (in the sense definied below) belong to the class of magnetic crystals characterized by a spontaneous parallel alignment of (atomic) magnetic moments below a certain temperature (the Curie temperature).

Ferrimagnetic are, for example, some compounds having the perovskite structure, *e.g.*, $FeN(Fe)_3$, $NiN(Fe)_3$ [1]. In the literature the term "ferrimagnet" is frequently used to describe a large class of magnetic crystals which differ from ferromagnets not only

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by different magnetic moments in the respective sublattices but also by the arbitrary sign of the Heisenberg exchange integral. Accordingly, the alignment of the magnetic moments (*i.e.*, effective atomic spins) in the (approximate) ground state may be either parallel or antiparallel (some authors include even materials with non-collinear spin configurations in the ground state). Since the latter group (*i.e.*, with antiparallel spin configuration) is also called "antiferrimagnets", we adopt Smart's [2] more precise terminology and use the term "ferrimagnets" exclusively for the first group to which the considerations in the present paper are restricted.

Although the theory of ferrimagnetism is based on the same principles as that of ferromagnetism, it is incomparably less developed. The first theoretical investigations of (in the broader sense) ferrimagnets are due to Néel [3] who applied the molecular field approach (cp. also [4, 5]). Later, ferrimagnetism has also been examined by other theoretical methods. By employing the quasiclassical Keffer-Kaplan-Yafet spin wave approach, it was analised by Cofta [6] who proved that the spin wave dispertion law has in the long-wavelength limit the form $\omega(k) \sim k^2$. The same result has been later obtained in [7] for the three-dimensional case. Similar results have been derived by Kubo [8], by somewhat different methods. Dyson's spin wave approach has been first applied to an isotropic ferrimagnet with external magnetic field by Kociński [9] who proved that, similarly as in ferromagnets, the dynamical interactions between spin waves can be neglected in the long-wavelength low-temperature limit. We may also mention that the spin wave method has been applied by Saeñz [10] and Wallace [11] to examine multi-sublattice magnetic systems with n magnetic ions in the unit cell, by taking into account in the Heisenberg spin Hamiltonian the interactions between all the magnetic ions of the unit cell themselves and the external magnetic field. The authors showed how the Hamiltonian can be diagonalized in the free-spin-waves approximation, and that it leads, generally, to a spin wave energy spectrum composed of n branches. A similar problem has also been solved by Kowalewski [12]. Dyson's spin wave theory has also been applied to two--sublattice ferrimagnets with cubic symmetry by Szaniecki [13] and Szweykowski [14]. In both papers, the corrections to thermodynamical quantities due to spin wave interactions have been calculated. The temperature-dependent Greens functions technique has been applied to two-sublattice isotropic ferrimagnets by Izyumow and Medvedev [15].

Recently [16], the thermodynamic properties of a ferromagnetic binary alloy with two atoms per unit cell (equivalent to a ferrimagnet) has been studied using the isotropic Heisenberg model, in order to determine the effect of the optical spin wave energy branch.

As is seen from the above survey, among the rather few papers devoted to ferrimagnetism only two considered anisotropic materials [6, 7], and even then in a quite phenomenological way (effective anisotropy field). Furthermore, in view of the recent refinements achieved in the spin wave theory of magnetic crystals [17–26] the case of ferrimagnetism certainly deserves new attention, particularly as regards the examination of the stable magnetic phases in the presence of an external magnetic field. A case in point is the newly discovered and extensively studied [27–34] second-order ferro-paramagnetic phase transition in uniaxial single-domain ferromagnets under the influence of a transversal magnetic field.

We thus examine a single-domain uniaxial two-sublattice Néel-type Heisenberg ferrimagnet with nearest-neighbour (inter-sublattice) exchange interactions, in the presence of a homogeneous external magnetic field. We consider in detail two extreme cases when the field is either parallel or perpendicular to the anisotropy axis (longitudinal and transversal field, respectively).

While restricting the considerations in the present paper to the free-spin-wave approximation (FSWA), in Part I the stable magnetic phases and the critical fields of the system are determined and the respective spin wave energy spectra examined. It is shown that in the transversal-field case the zero-temperature critical field for the "ferri"-paramagnetic phase transition as determined from the spin wave energy spectrum coincides with that obtained in a former paper [22] from the stability conditions for the system's approximate ground state. In Part II, the low-temperature thermodynamics of the system in the FSWA is given, while the influence of spin wave interactions will be considered in a separate paper [35].

It appears that a positive uniaxial anisotropy (magnetically preferred plane) leads to qualitatively different results. Hence, this problem is also deferred to a subsequent paper [36].

2. *The Hamiltonian*

We assume the following spin Hamiltonian for our system:

$$\mathscr{H} = -I \sum_{\langle f,g \rangle} (\tilde{S}_f^x \tilde{S}_g^x + \tilde{S}_f^y \tilde{S}_g^y + Z \tilde{S}_f^z \tilde{S}_g^z) - \mu_1 \boldsymbol{H} \sum_f \tilde{\boldsymbol{S}}_f - \mu_2 \boldsymbol{H} \sum_g \tilde{\boldsymbol{S}}_g \tag{1}$$

where I is the (positive) nearest-neighbour exchange integral, $Z = K/I$ with the anisotropy constant K assumed to be positive, $\mu_n = g_n \mu$ is the effective atomic magnetic moment in the n-th sublattice ($n = 1, 2$), g_n are Landé's splitting factors, μ denotes Bohr's magneton

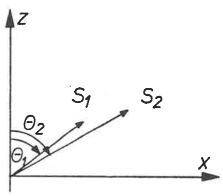

Fig. 1

and $\boldsymbol{H}$ the external magnetic field. The subscripts f, g denote lattice vectors in the first and second sublattice, respectively.

Taking into account the spatial symmetry of the Hamiltonian (1) we may, without loss of generality, put $H_y = 0$.

Similar as in [22], we introduce in each sublattice a local coordinate system whose

z-axis is the direction of spin quantization. We perform the following rotations of the spins in the plane $x0z$ (Fig. 1):

$$\begin{aligned} \tilde{S}_f^x &= S_f^x \cos\theta_1 + S_f^z \sin\theta_1 & \tilde{S}_g^x &= S_g^x \cos\theta_2 + S_g^z \sin\theta_2 \\ \tilde{S}_f^y &= S_f^y & \tilde{S}_g^y &= S_g^y \\ \tilde{S}_f^z &= -S_f^x \sin\theta_1 + S_f^z \cos\theta_1 & \tilde{S}_g^z &= -S_g^x \sin\theta_2 + S_g^z \cos\theta_2. \end{aligned} \tag{2}$$

With the spin deviation operators

$$S_{f,g}^{\pm} = S_{f,g}^x \pm i S_{f,g}^y \tag{3}$$

the Hamiltonian (1) takes upon the transformation (2) the form

$$\begin{aligned} \mathscr{H} = -I \sum_{\langle f,g \rangle} \{ & S_f^z S_g^z (Z \cos\theta_1 \cos\theta_2 + \sin\theta_1 \sin\theta_2) + \tfrac{1}{2}(S_f^+ S_g^z + S_f^- S_g^z) \times \\ & \times (\cos\theta_1 \sin\theta_2 - Z \sin\theta_1 \cos\theta_2) + \tfrac{1}{2}(S_f^z S_g^+ + S_f^z S_g^-)(\sin\theta_1 \cos\theta_2 - Z \cos\theta_1 \sin\theta_2) + \\ & + \tfrac{1}{4}(S_f^+ S_g^+ + S_f^- S_g^-)(Z \sin\theta_1 \sin\theta_2 + \cos\theta_1 \cos\theta_2 - 1) + \tfrac{1}{4}(S_f^- S_g^+ + S_f^+ S_g^-) \times \\ & \times (Z \sin\theta_1 \sin\theta_2 + \cos\theta_1 \cos\theta_2 + 1) - \mu_1 \{ H_x \sum_f [S_f^z \sin\theta_1 + \tfrac{1}{2}(S_f^+ + S_f^-) \cos\theta_1] - \\ & - H_z \sum_f [S_f^z \cos\theta_1 - \tfrac{1}{2}(S_f^+ + S_f^-) \sin\theta_1] \} - \mu_2 \{ H_x \sum_g [S_g^z \sin\theta_2 + \\ & + \tfrac{1}{2}(S_g^+ + S_g^-) \cos\theta_2] - H_z \sum_g [S_g^z \cos\theta_2 - \tfrac{1}{2}(S_g^+ + S_g^-) \sin\theta_2] \}. \end{aligned} \tag{4}$$

We pass to the Bose representation by means of the Holstein-Primakoff mapping [37]:

$$\begin{aligned} (S_f^-)^+ &= S_f^+ = (2S_1)^{\frac{1}{2}} \varphi(n_f) a_f, & S_f^z &= S_1 - n_f; \\ (S_g^-)^+ &= S_g^+ = (2S_2)^{\frac{1}{2}} \varphi(n_g) b_g, & S_g^z &= S_2 - n_g \end{aligned} \tag{5}$$

where

$$\begin{aligned} \varphi(n_f) = \left(1 - \frac{n_f}{2S_1}\right)^{\frac{1}{2}}, \quad \varphi(n_g) = \left(1 - \frac{n_g}{2S_2}\right)^{\frac{1}{2}}; \\ n_f = a_f^+ a_f, \quad n_g = b_g^+ b_g \end{aligned} \tag{6}$$

and

$$[a_f, a_{f'}^+] = \delta_{ff'}, \quad [b_g, b_{g'}^+] = \delta_{gg'}. \tag{7}$$

The commutators of other combinations of Bose operators are equal to zero.

Substituting (5) into (4) and expanding (6) in a power series,

$$\varphi(n_f) = 1 - \frac{n_f}{4S_1} - \ldots, \quad \varphi(n_g) = 1 - \frac{n_g}{4S_2} - \ldots, \tag{8}$$

we can write the transformed Hamiltonian in the form

$$\mathscr{H} = \mathscr{H}_0+\mathscr{H}_1+\mathscr{H}_2+\dots \tag{9}$$

where $\mathscr{H}_n$ is a linear combination of products of n Bose operators.

Let us introduce the Fourier transform of a_f and b_g:

$$a_f = N^{-\frac{1}{2}}\sum_k e^{ikf}a_k, \quad b_g = N^{-\frac{1}{2}}\sum_k e^{ikg}b_k \tag{10}$$

where k are wave vectors and N is the number of (magnetic) sublattice sites. Then, we obtain for $\mathscr{H}_0$, $\mathscr{H}_1$, $\mathscr{H}_2$:

$$\mathscr{H}_0 = -d(Z\cos\theta_1\cos\theta_2+\sin\theta_1\sin\theta_2+h_x\sin\theta_1+h_z\cos\theta_1+ \\ +\kappa h_x\sin\theta_2+\kappa k_z\cos\theta_2) \tag{11}$$

where
$d = \bar{I}/NS_1S_2$, $\kappa = \mu_2S_2/\mu_1S_1$, $h_{x,z} = \mu_1H_{x,z}/S_2\bar{I}$, $\bar{I} = zI$ with z as the number of nearest neighbours;

$$\mathscr{H}_1 = A_0(a_0+a_0^+)+B_0(b_0+b_0^+) \tag{12}$$

where

$$A_0 = \tfrac{1}{2}(2NS_1)^{\frac{1}{2}}\bar{I}S_2(Z\sin\theta_1\cos\theta_2-\cos\theta_1\sin\theta_2-h_x\cos\theta_1+h_z\sin\theta_1),$$
$$B_0 = \tfrac{1}{2}(2NS_2)^{\frac{1}{2}}\bar{I}S_1(Z\cos\theta_1\sin\theta_2-\cos\theta_2\sin\theta_1-\kappa h_x\cos\theta_2+\kappa h_z\sin\theta_2); \tag{13}$$

and

$$\mathscr{H}_2 = m_1\sum_k a_k^+a_k+m_2\sum_k b_k^+b_k+m_3\sum_k\gamma_k(a_k^+b_k+a_kb_k^+)+ \\ +m_4\sum_k\gamma_k(a_k^+b_{-k}^++a_kb_{-k}) \tag{14}$$

where

$$m_1 = \bar{I}S_2(Z\cos\theta_1\cos\theta_2+\sin\theta_1\sin\theta_2+h_x\sin\theta_1+h_z\cos\theta_1),$$
$$m_2 = \bar{I}S_1(Z\cos\theta_1\cos\theta_2+\sin\theta_1\sin\theta_2+\kappa h_x\sin\theta_2+\kappa h_z\cos\theta_2),$$
$$m_3 = -\tfrac{1}{2}\bar{I}(S_1S_2)^{\frac{1}{2}}(Z\sin\theta_1\sin\theta_2+\cos\theta_1\cos\theta_2+1),$$
$$m_4 = -\tfrac{1}{2}\bar{I}(S_2S_1)^{\frac{1}{2}}(Z\sin\theta_1\sin\theta_2+\cos\theta_1\cos\theta_2-1). \tag{15}$$

3. The stable magnetic phases and critical fields

The angles θ_1 and θ_2 are determined from the necessary minimum conditions

$$\frac{\partial\tilde{\mathscr{H}}_0}{\partial\theta_1} = \frac{\partial\tilde{\mathscr{H}}_0}{\partial\theta_2} = 0, \quad \tilde{\mathscr{H}}_0 \equiv \mathscr{H}_0/NS_1S_2\bar{I} \tag{16}$$

which have the following field-dependent solutions:

I. For the case $h_z \neq 0$, $h_x = 0$ (field parallel to the easy axis) we obtain:

1) $\sin\theta_1 = \sin\theta_2 = 0,$ (17)

2) $\cos\theta_1 = (R' + \kappa Z)h_z/|w|, \; \cos\theta_2 = (\kappa R'^{-1} + Z)h_z/|w|$ (18)

where $R' = \left(\dfrac{|w| + \kappa^2 h_z^2}{|w| + h_z^2}\right)^{\frac{1}{2}}$ and $w = Z^2 - 1$.

From the sufficient conditions

$$\Delta = \frac{\partial^2 \tilde{\mathscr{H}}_0}{\partial\theta_1\partial\theta_1} \cdot \frac{\partial^2 \tilde{\mathscr{H}}_0}{\partial\theta_2\partial\theta_2} - \left(\frac{\partial^2 \tilde{\mathscr{H}}_0}{\partial\theta_1\partial\theta_2}\right)^2 > 0, \qquad \frac{\partial^2 \tilde{\mathscr{H}}_0}{\partial\theta_1\partial\theta_1} > 0 \tag{19}$$

it follows that in the case $K > 0$ the solution (17) corresponds to a minimum of $\mathscr{H}_0$ and that

$$\text{a) } \theta_1 = \theta_2 = 0 \text{ for } h_z > \frac{-Z(\kappa+1) + \sqrt{Z^2(\kappa+1)^2 - 4\kappa|w|}}{2\kappa} \equiv -h_{zc} \tag{20a}$$

$$\text{b) } \theta_1 = \theta_2 = \pi \text{ for } h_z < h_{zc}. \tag{20b}$$

Hence, for $|h_z| > h_{zc}$ the spins are always parallel to the field whereas for $|h_z| < h_{zc}$ they can be either parallel or antiparallel to it.

For the (approximate) ground state energy we have:

a) $\tilde{\mathscr{H}}_0 = -Z - (\kappa+1)h_z,$ (21a)

b) $\tilde{\mathscr{H}}_0 = -Z + (\kappa+1)h_z.$ (21b)

This field dependence is illustrated in Fig. 2.

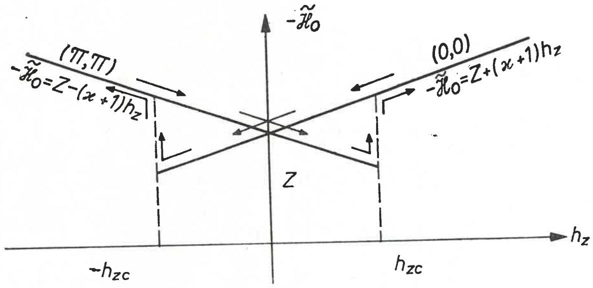

Fig. 2

II. For the case $h_z = 0$, $h_x \neq 0$ (field perpendicular to the easy axis) we have, according to [22], the following solutions of Eqs. (16):

1) $\sin\theta_1 = (ZR + \kappa)h_x/w, \; \sin\theta_2 = (\kappa Z/R + 1)h_x/w;$ (22a, b)

$\cos\theta_1 = R\cos\theta_2$ (23)

where

$$R = \left(\frac{w+\kappa^2 h_x^2}{w+h_x^2}\right)^{\frac{1}{2}}; \qquad w = Z^2-1 \tag{24}$$

and

$$\text{2)} \quad \cos\theta_1 = \cos\theta_2 = 0. \tag{25}$$

It was shown in [22] that the solution 1) corresponds to a minimum of $\mathscr{H}_0$ for $|h_x| < h_{xc}$ where

$$h_{xc} = \frac{\sqrt{(\kappa+1)^2+4\kappa w}-(\kappa+1)}{2\kappa}. \tag{26}$$

This solution describes a spin configuration that we shall call henceforth "scissor phase" (SP) (see Fig. 1). It was also shown in [22] that the solution (25) corresponds to a minimum of $\mathscr{H}_0$ for $|h_x| > h_{xc}$. In this case all the spins are aligned along the field which corresponds to a paramagnetic phase (PP).

The dependence of the (approximate) ground state energy on the field strength was given and illustrated in [22] for both phases, SP, and PP.

4. The spin wave energy spectra

The bilinear part $\mathscr{H}_2$ of the Hamiltonian can be diagonalized [21, 38] by means of the following canonical transformation:

$$\begin{aligned} a_k &= u_{11}(k)\alpha_k + v_{11}(k)\alpha_{-k}^+ + u_{12}(k)\beta_k + v_{12}(k)\beta_{-k}^+, \\ b_k &= u_{21}(k)\alpha_k + v_{21}(k)\alpha_{-k}^+ + u_{22}(k)\beta_k + v_{22}(k)\beta_{-k}^+, \end{aligned} \tag{27}$$

and $\mathscr{H}_2$ takes the form

$$\mathscr{H}_2 = \sum_k (E_k^{(1)}\alpha_k^+\alpha_k + E_k^{(2)}\beta_k^+\beta_k) + \Delta\mathscr{H}_2 \tag{28}$$

where

$$\Delta\mathscr{H}_2 = -\sum_{k,n} [v_{1n}^2(k) + v_{2n}^2(k)] E_k^{(n)}; \qquad n \equiv 1, 2. \tag{29}$$

The coefficients (u, v) are determined from the set of equations

$$\begin{pmatrix} m_1 - E_k^{(n)} & m_3\gamma_k & 0 & m_4\gamma_k \\ m_3\gamma_k & m_2 - E_k^{(n)} & m_4\gamma_k & 0 \\ 0 & m_4\gamma_k & m_1 + E_k^{(n)} & m_3\gamma_k \\ m_4\gamma_k & 0 & m_3\gamma_k & m_2 + E_k^{(n)} \end{pmatrix} \cdot \begin{pmatrix} u_{1n} \\ u_{2n} \\ v_{1n} \\ v_{2n} \end{pmatrix} = 0 \tag{30}$$

where $\gamma_k = z^{-1} \sum_\delta e^{ik\delta}$, with δ as the vector to the nearest neighbour.

These equations have nontrivial solutions if the determinant vanishes, which lead to the spin wave spectra (cp. [21])

$$(E_k^{(n)})^2 = \tfrac{1}{2}(m_1^2+m_2^2)+(m_3^2-m_4^2)\gamma_k^2 \pm$$
$$\pm\{\tfrac{1}{4}(m_1^2-m_2^2)^2+[(m_1+m_2)^2m_3^2-(m_1-m_2)^2m_4^2]\gamma_k^2\}^{\frac{1}{2}} \tag{31}$$

where the sign $\pm$ corresponds to $n = 1, 2$.

When determining from Eqs (30) the coefficients (u, v) corresponding to the solutions (31), one must take into account the canonical conditions of the transformation (27) which read

$$\sum_{n/1}^{2}(u_{1n}u_{2n}-v_{1n}v_{2n}) = 0, \quad \sum_{n/1}^{2}(u_{1n}v_{2n}-u_{2n}v_{1n}) = 0,$$
$$\sum_{n/1}^{2}(u_{mn}^2-v_{mn}^2) = 1,$$
$$\sum_{n/1}^{2}(u_{n1}v_{n2}-u_{n2}v_{n1}) = 0, \quad \sum_{n/1}^{2}(u_{n1}u_{n2}-v_{n1}v_{n2}) = 0,$$
$$\sum_{n/1}^{2}(u_{nm}^2-v_{nm}^2) = 1, \qquad m \equiv 1, 2. \tag{32}$$

The solutions are given in the Appendix.

In closer examining the energy spectra (31) we shall specify the direction of the magnetic field.

I. The case $h_x = 0$, $h_z \neq 0$ (longitudinal field).

In this case $m_4 = 0$ and the formula (31) reduces to

$$(E_k^{(n)})^2 = \tfrac{1}{4}[(m_1+m_2)\pm\sqrt{(m_1-m_2)^2+m_3^2\gamma_k^2}]^2. \tag{33}$$

In the other hand, the set of equations (30) splits into two subsets,

$$\begin{pmatrix} m_1-E_k^{(n)} & m_3\gamma_k \\ m_3\gamma_k & m_2-E_k^{(n)} \end{pmatrix}\cdot\begin{pmatrix} u_{1n} \\ u_{2n} \end{pmatrix} = 0, \quad \begin{pmatrix} m_1+E_k^{(n)} & m_3\gamma_k \\ m_3\gamma_k & m_2+E_k^{(n)} \end{pmatrix}\cdot\begin{pmatrix} v_{1n} \\ v_{2n} \end{pmatrix} = 0. \tag{34}$$

From the first subset we obtain the positive energy branches

$$E_k^{1,2} \equiv E_k^{\pm} = \tfrac{1}{2}[(m_1+m_2)\pm\sqrt{(m_1-m_2)^2+4m_3^2\gamma_k^2}] \tag{35}$$

for which the coefficients of the diagonalizing transformation have the form

$$u_{11} = \frac{m_3\gamma_k}{[(m_1-E_k^-)^2+m_3^2\gamma_k^2]^{\frac{1}{2}}}, \quad u_{12} = \frac{m_4\gamma_k}{[(m_1-E_k^+)^2+m_3^2\gamma_k^2]^{\frac{1}{2}}}, \tag{36}$$
$$u_{21} = \frac{-(m_1-E_k^-)}{[(m_1-E_k^-)^2+m_3^2\gamma_k^2]^{\frac{1}{2}}}, \quad u_{22} = \frac{-(m_1-E_k^+)}{[(m_1-E_k^+)^2+m_3^2\gamma_k^2]^{\frac{1}{2}}},$$
$$v_{nm} = 0, \qquad (n, m \equiv 1, 2).$$

Taking into account Eqs (15) and (20a, b) we obtain

$$m_1 = \bar{I}S_2(Z \pm h_z), \quad m_2 = \bar{I}S_1(Z \pm \kappa h_z),$$

$$m_3 = -\bar{I}(S_1 S_2)^{\frac{1}{2}}, \quad m_4 = 0 \tag{37}$$

where the upper sign corresponds to the case "a", Eq. (20a). Hence, with the notation $\tilde{S} = S_2/S_1$ we have

$$\text{a)} \quad E_k^{\pm} = \tfrac{1}{2}\bar{I}S_1\{Z(\tilde{S}+1)+(\tilde{S}+\kappa)h_z \pm \sqrt{[Z(\tilde{S}-1)+(\tilde{S}-\kappa h_z)]^2+4\tilde{S}\gamma_k^2}\} \tag{38a}$$

for $h_z > -h_{zc}$, and

$$\text{b)} \quad E_k^{\pm} = \tfrac{1}{2}\bar{I}S_1\{Z(\tilde{S}+1)-(\tilde{S}+\kappa)h_z \pm \sqrt{[Z(\tilde{S}-1)-(\tilde{S}-\kappa h_z)]^2+4\tilde{S}\gamma_k^2}\} \tag{38b}$$

for $h_z < h_{zc}$.

II. The case $h_x \neq 0$, $h_z = 0$ (transversal field).

The formula (31) may be written in the form

$$E_k^{\mp} = (a+b\gamma_k^2 \mp \sqrt{c+d\gamma_k^2})^{\frac{1}{2}} \tag{39}$$

where

$$a = \tfrac{1}{2}(m_1^2+m_2^2), \quad b = m_3^2-m_4^2, \quad c = \tfrac{1}{4}(m_1^2-m_2^2)^2,$$

$$d = (m_1^2+m_2^2)m_3^2-(m_1^2-m_2^2)^2 m_4^2. \tag{40}$$

In particular, we have

1) for the scissor phase (SP):

$$a = \tfrac{1}{2}\bar{I}^2 S_1^2 Z^2(\tilde{S}^2 R^{-2}+R^2), \quad b = \bar{I}^2 S_1^2 \tilde{S}[R+(\kappa Z+R)h_x^2/w],$$

$$c = \tfrac{1}{4}\bar{I}^4 \tilde{S}_1^4 Z^4(\tilde{S}^2 R^{-2}-R^2), \quad d = \bar{I}^4 S_1^4 \tilde{S} Z^2(AR+\tilde{S}R^{-1})\cdot(A\tilde{S}R^{-1}+R) \tag{41}$$

where

$$A = R+(\kappa Z+R)h_x^2/w;$$

2) for the paramagnetic phase (PP):

$$a = \tfrac{1}{2}\bar{I}^2 S_1^2[\tilde{S}^2(1+h_x)^2+(1+\kappa h_x)^2], \quad b = \bar{I}^2 S_1 S_2 Z,$$

$$c = \tfrac{1}{4}\bar{I}^4 S_1^4[\tilde{S}^2(1+h_x)^2-(1+\kappa h_x)^2],$$

$$d = \bar{I}^4 S_1^4 \tilde{S}[\tilde{S}(1+h_x)+Z(1+\kappa h_x)]\cdot[Z\tilde{S}(1+h_x)+1+\kappa h_x]. \tag{42}$$

It remains to be proven that the above energy spectra are real and non-negative in the stability intervals for the respective magnetic phases following from Eqs (20) and (26), *i. e.*, we must show that the inequality

$$E_k^{\pm} \geqslant 0 \tag{43}$$

holds for all vectors k.

I. The case $h_x = 0$, $h_z \neq 0$

In this case E_k is given by Eqs (38) for which the inequality (43) leads to the conditions:

$$\text{a)}\quad h_z \geqslant \frac{-Z(\kappa+1)+\sqrt{Z^2(\kappa+1)^2-4\kappa(Z^2-\gamma_k^2)}}{2\kappa} = -h_{zc}(k)$$

or

$$h_z \leqslant \frac{-Z(\kappa+1)-\sqrt{Z^2(\kappa+1)^2-4\kappa(Z^2-\gamma_k^2)}}{2\kappa} = -h'_{zc}(k); \tag{44a}$$

$$\text{b)}\quad h_z \leqslant h_{zc}(k)$$

or

$$h_z \geqslant h'_{zc}(k). \tag{44b}$$

We easily verify that the first of the above conditions in each case, a) and b), will ensure inequality (43) to be satisfied for all k if we put $k = 0$ in $h_{zc}(k)$. In that case, however, we readily conclude from Eqs (20a) and (44a) that $h_{zc}(0) = h_{zc}$.

II. The case $h_z = 0$, $h_x \neq 0$

For convenience we rewrite the energy (39) in this case in the form

$$(E_k^{\pm})^2 = P \pm (P^2-Q)^{\frac{1}{2}} \tag{45}$$

where

$$P = \tfrac{1}{2}(m_1^2+m_2^2)+(m_3^2-m_4^2)\gamma_k^2 > 0,$$

$$Q = [(m_3-m_4)^2\gamma_k^2-m_1m_2]\,[(m_3+m_4)^2\gamma_k^2-m_1m_2]. \tag{46}$$

The inequality (43) is equivalent to

$$P^2-Q \geqslant 0 \text{ and } Q \geqslant 0. \tag{47}$$

Because of

$$P^2-Q = \tfrac{1}{4}(m_1^2-m_2^2)^2+(m_1^2+m_2^2)(m_3^2-m_4^2)\gamma_k^2+2m_1m_2(m_3^2+m_4^2)\gamma_k^2,$$

$$\gamma_k^2(m_3^2-m_4^2) \geqslant 0, \quad m_1m_2 > 0, \tag{48}$$

the first of the inequalities (47) is always true. The second one we easily prove to hold for all k as follows:

1) in the "scissor phase" (SP) we have from Eqs (26) and (41)

$$Q = (\bar{I}^2S_1S_2)^2(Z^2-\gamma_k^2)(Z^2-A^2\gamma_k^2) \geqslant 0 \text{ for } 0 \leqslant h_x < h_{xc}$$

because of $1 \leqslant A \leqslant Z$ and $0 \leqslant \gamma_k^2 \leqslant 1$;

2) in the paramagnetic phase (PP) we obtain from Eq. (42)

$$Q = (\bar{I}^2S_1S_2)^2[(1+h_x)(1+\kappa h_x)-\gamma_k^2]\,[(1+h_x)(1+\kappa h_x)-Z^2\gamma_k^2].$$

The quantity in the first square brackets is always positive, and that in the second ones for

$$|h_x| > \frac{\sqrt{(\kappa+1)^2+4\kappa(Z^2\gamma_k^2-1)}-(\kappa+1)}{2\kappa} \equiv h_{xc}(k). \tag{49}$$

Hence, the inequality $Q \geqslant 0$ will be fulfilled for all k if we put $h_{xc}(0)$ in (49), and from Eq. (26) we see that $h_{xc}(0) = h_{xc}$.

5. *Final remarks*

The results concerning the system's ground state in the longitudinal field case given here supplement those obtained for the transversal-field case in [22]. It is seen that in the longitudinal-field case there exist, strictly speaking, two stable magnetic phases which overlap in the field-interval $\langle -h_{zc}, h_{zc} \rangle$ in which one of them is actually meta-stable (cp. Fig. 2). However, if the system is in the meta-stable phase (spins antiparallel to the magnetic field) it cannot pass by itself to the energetically lower stable state (with spins parallel to the field), as this is prevented by an energy barrier due to the uniaxial anisotropy (the spins would have to pass through the magnetically hard plane). This leads to the well-known rectangular hysteresis loop for the system's zero-temperature total magnetization M_0 shown in Fig. 3. Note that the phase transition at $\pm h_{zc}$ is actually of zeroth

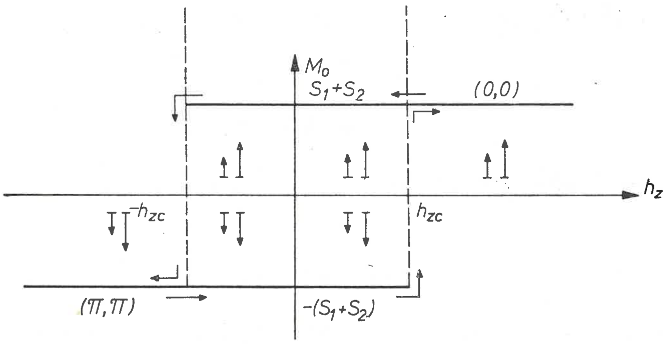

Fig. 3

order (discontinuity of the energy), quite like the transition from the AF to the SF phase in antiferromagnets [24], in contrast to the phase transition SP → PP in the transversal-field case which has been shown in [22] to be of second order.

The main result of the present paper is the derivation (in the FSWA) of the spin wave energy spectrum (31) for an arbitrary direction of the external magnetic field, and the proof that in the longitudinal as well in the transversal-field case a careful analysis of the positiveness and reality of the energy spectra leads to the same field intervals as the minimization of the system's approximate ground state energy. These results are utilized in Part II in studying the system's thermodynamic behaviour and will be extended in [36] to the case of uniaxial two-sublattice ferromagnets with a magnetically preferred plane.

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APPENDIX

The coefficients $u_{11}(k)$, $v_{11}(k)$, ... from Eq. (30) have the following form [21]:

$$u_{1n} = \frac{X_{1n}}{\Delta} v_{1n}, \quad u_{2n} = \frac{X_{2n}}{\Delta} v_{1n}, \quad v_{2n} = \frac{X_{3n}}{\Delta} v_{1n},$$

$$v_{11}^2 = \frac{\Delta^2(X_{12}X_{22} - X_{32}\Delta)}{B}, \quad v_{12}^2 = \frac{\Delta^2(X_{31}\Delta - X_{11}X_{21})}{B}$$

where

$$X_{1n} = -(m_1 + E^{(n)})\tfrac{1}{2}[m_1^2 - m_2^2 - (-1)^n A] + [m_4^2(m_1 - m_2) - m_3^2(m_1 + m_2)]\gamma_k^2$$

$$X_{2n} = -\gamma_k\{E^{(n)}(m_1 + m_2) + \tfrac{1}{2}[(m_1 + m_2)^2 - (-1)^n A]\}m_3,$$

$$X_{3n} = m_4\gamma_k\{\tfrac{1}{2}(m_1 - m_2)^2 + E^{(n)}(m_1 - m_2) - (-1)^n \tfrac{1}{2} A\},$$

$$B = (X_{11}^2 - \Delta^2)(X_{12}X_{22} - X_{32}\Delta) - (X_{12}^2 - \Delta^2)(X_{11}X_{21} - X_{31}\Delta), \quad (n \equiv 1, 2)$$

and

$$\Delta = 2m_1 m_3 m_4 \gamma_k^2$$

$$A = \{(m_1^2 - m_2^2)^2 + 4[(m_1 + m_2)^2 m_3^2 - (m_1 - m_2)^2 m_4^2]\gamma_k^2\}^{\frac{1}{2}}.$$

REFERENCES

[1] J. B. Goodenough, J. M. Longo, in *Landolt-Börnstein*, vol. IV/a, p. 126, Springer-Verlag, Berlin-New York 1970.
[2] J. S. Smart, *Amer. J. Phys.*, **23**, 356 (1955).
[3] L. Néel, *Ann. Phys.* (France), **3**, 137 (1948).
[4] J. S. Smart, *Effective Field Theories of Magnetism*, W. B. Saunders Co, Philadelphia–London 1966.
[5] A. H. Morrish, *The Physical Principles of Magnetism*, John Wiley and Sons Inc., New York 1965.
[6] H. Cofta, *Acta Phys. Polon.*, **15**, 311 (1956).
[7] H. Cofta, *Acta Phys. Polon.*, **18**, 215 (1959).
[8] R. Kubo, *Advances in Phys.*, **4** (1955).
[9] J. Kociński, *Acta Phys. Polon.*, **29**, 565 (1960).
[10] A. W. Saenz, *Phys. Rev.*, **125**, 1940 (1962).
[11] D. C. Wallace, *Phys. Rev.*, **128**, 1614 (1962).
[12] L. Kowalewski, *Acta Phys. Polon.*, **34**, 415 (1963).
[13] J. Szaniecki, *Acta Phys. Polon.*, **26**, 37 (1964).
[14] P. Szweykowski, *Acta Phys. Polon.*, **30**, 415 (1966).

[15] W. Izyumov, W. Medvedev, *Theory of Magnetic Ordered Crystals with Impurities*, Moscow 1970 (in Russian).
[16] F. Leoni, *preprint No RTI/FNA*, C.N.E.N., Roma 1969.
[17] W. J. Ziętek, *Acta Phys. Polon.*, **35**, 799 (1969); **36**, 145 (1969).
[18] H. Pfeiffer, W. J. Ziętek, *Acta Phys. Polon.*, **A38**, 205 (1970).
[19] L. Biegała, *Acta Phys. Polon.*, **A38**, 773 (1970).
[20] H. Pfeiffer, *Acta Phys. Polon.*, **A39**, 209 (1971).
[21] A. Pękalski, W. J. Ziętek, *Acta Phys. Polon.*, **A39**, 327 (1971).
[22] G. Kozłowski, L. Biegała, S. Krzemiński, *Acta Phys. Polon.*, **A39**, 417 (1971).
[23] S. Krzemiński, *Acta Phys. Polon.*, **A39**, 201, 661 (1971).
[24] A. Pękalski, *Acta Phys. Polon.*, **A40**, 189 (1971).
[25] H. Pfeiffer, *Acta Phys. Polon.*, **A40**, 567 (1971).
[26] J. Ulner, *Acta Phys. Polon.*, **A40**, 725 (1971).
[27] P. Woytowicz, M. Rayl, *Phys. Rev. Letters*, **20**, 1489 (1968).
[28] E. Riedel, F. Wegner, *Z. Phys.*, **225**, 195 (1969).
[29] H. Thomas, *Phys. Rev.*, **187**, 630 (1969).
[30] K. Durczewski, *Acta Phys. Polon.*, **A38**, 855 (1970).
[31] J. Sznajd, *Phys. Status Solidi*, **41**, 405 (1970).
[32] H. Pfeiffer, *Acta Phys. Polon.*, **A39**, 213 (1971).
[33] H. Suzuki, T. Watanabe, *J. Phys. Soc. Japan*, **30**, 367 (1971).
[34] W. Wasilewski, W. J. Ziętek, *Acta Phys. Polon.*, **A42**, 207 (1972).
[35] L. Biegała, W. J. Ziętek, to be published.
[36] L. Biegała, to be published.
[37] T. Holstein, H. Primakoff, *Phys. Rev.*, **58**, 1096 (1940).
[38] S. V. Tyablikov, *Methods of the Quantum Theory of Magnetism*, Moscow 1965 (in Russian).